LA-UR-21-22766
Approved for public release; distribution is unlimited.

Title: DFT for design and characterization of functional materials

Author(s): Nelson, Tammie Renee

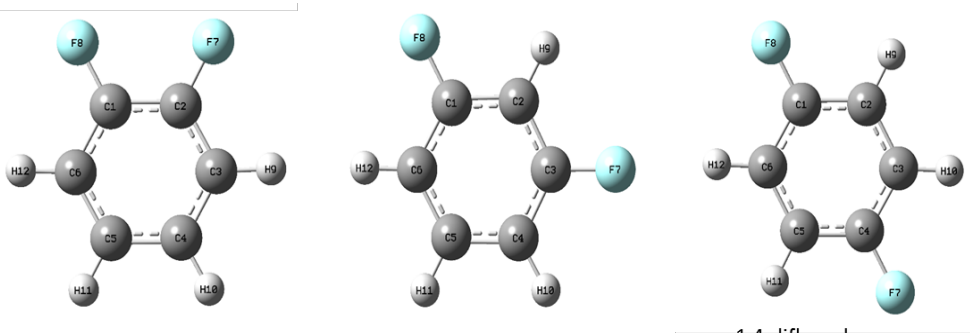
Intended for: Report

Issued: 2021-03-22

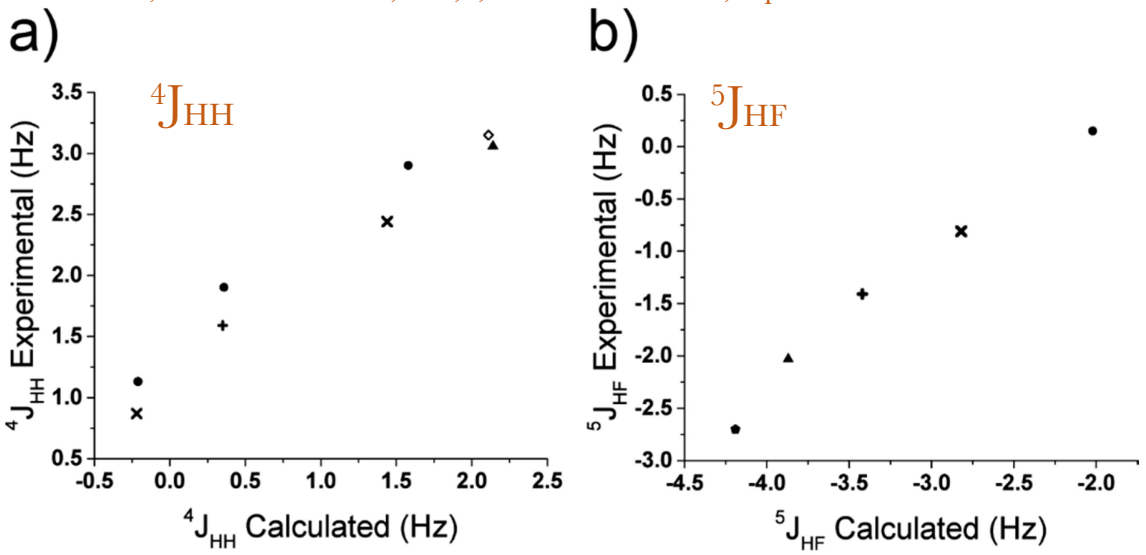
DFT for design and characterization of functional materials

Simulations supporting development of J-coupled spectroscopy using density functional theory (DFT) Gaussian 16 software package supporting LDRD DR and ECR

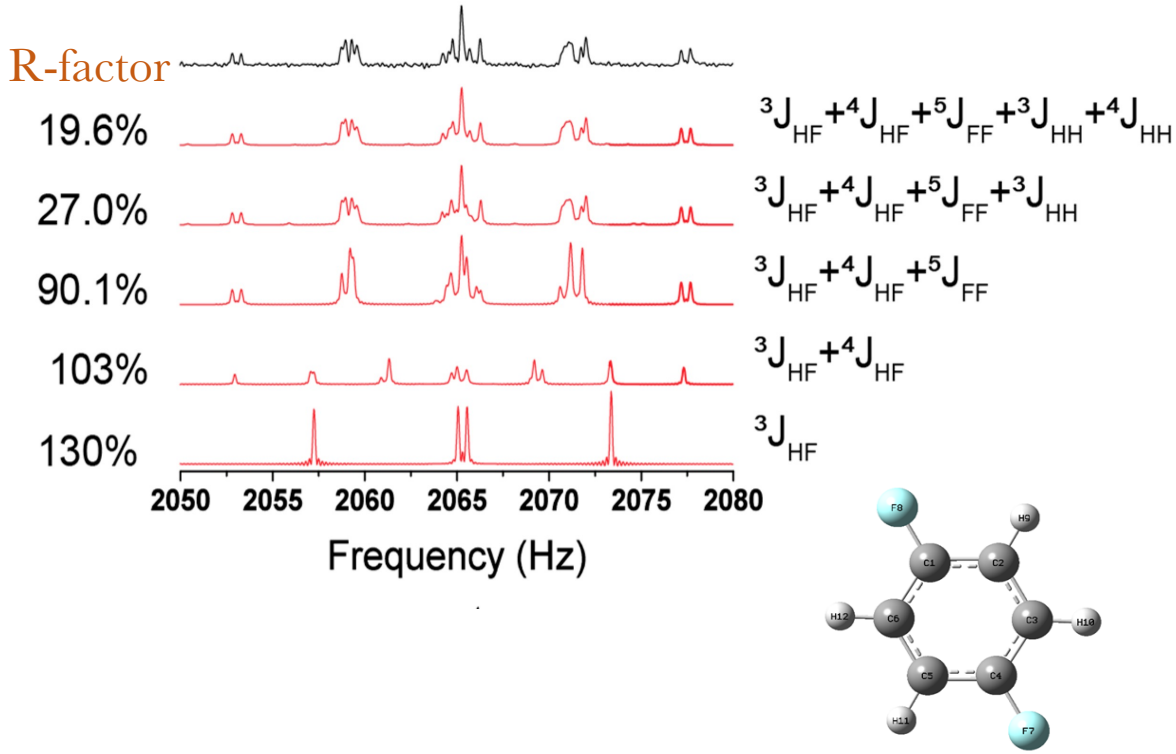
Representative difluorobenzene isomers



●monofluorobenzene; +1,2-difluorobenzene; ×1,3-difluorobenzene;
◇1,4-difluorobenzene; ▲1,2,4-trifluorobenzene; ⬟pentafluorobenzene



Comparison between experimental and ab initio calculated. When J-couplings are near zero, a sign error may result. This is remedied by applying a linear correction factor.



The simulated (red) and experimental spectra (black) of the ^{19}F region of 1,4-difluorobenzene. The simulated spectra each contain an additional J-coupling. Clearly, the subspectra are well simulated only when all heteronuclear and homonuclear J-couplings were included.

DFT for design and characterization of functional materials

Develop a modeling framework to predict IR spectra of glassy materials using VASP software package supporting program development

Oscillator strengths for IR active modes:

$$f(\nu) = \sum_{\alpha} \left| \sum_{s\beta} Z^*_{\alpha\beta}(s) e_{\beta}(s|\nu) \right|^2$$

$e_{\beta}(s|n)$ normalized vibrational eigenvector of mode ν

$Z^*_{\alpha\beta}$ Born effective-charge tensor of atom s

Task 1 compute **(1)** and **(2)** for an optimized geometry using VASP

Unit cell geometry → optimization → Optimized unit cell geometry (*If forces $F_i \approx 0$; *If forces $F_i \neq 0$) → Supercell 2x2x2 → optimization → Optimized supercell (*If forces $F_i \approx 0$; *If forces $F_i \neq 0$) → Phonon calculation

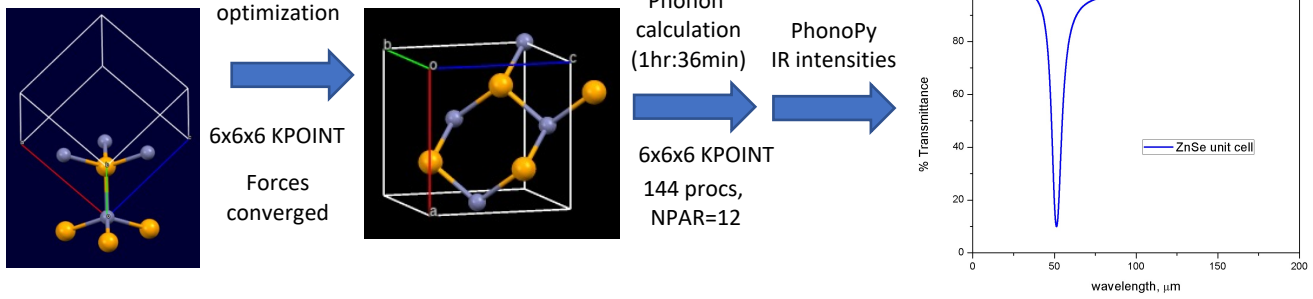
Task 2

Phonon modes & BEC tensor → IR and Raman spectra

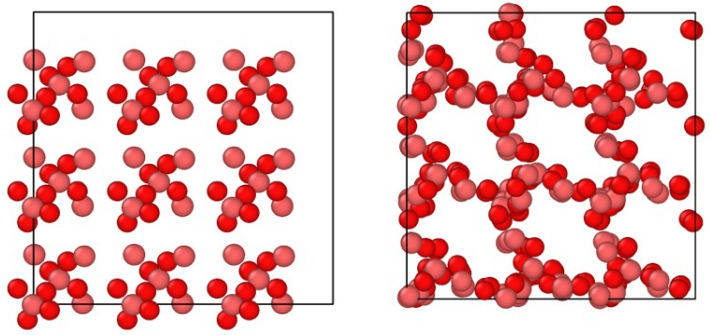
PhonoPy-SpectroscoPy package to post process data and compute IR intensities

(1) Phonon modes from DFPT (Density-Functional Perturbation Theory) method available in VASP 5.* version

(2) Born effective charge (BEC) tensor giving the change of atoms polarizability w.r.t. an external electric field



Workflow and simulated IR transmission spectrum for ZnSe simulation cell with 8 atoms



Preliminary result for generating initial amorphous silicon SiO_2 geometries starting from a supercell at 0K (left) and using NVT heating to 4000K (right).